



Cathode refunctionalization as a lithium ion battery recycling alternative



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HIGHLIGHTS

- Commercial LiFePO₄ cells were cycled to an end-of-life (EOL) state.
- Capacity fade with cycling was determined to be due to lithium loss.
- LiFePO₄ capacity was recovered through electrochemical and chemical re-lithiation.
- Re-lithiation is a process to refunctionalize cathode to regain performance.
- Refunctionalization has less embodied energy than material synthesis or recycling.

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ABSTRACT

An approach to battery end-of-life (EOL) management is developed involving cathode refunctionalization, which enables remanufacturing of the cathode from EOL materials to regain the electrochemical performance. To date, the optimal end-of-life management of cathode materials is based on economic value and environmental impact which can influence the methods and stage of recycling. Traditional recycling methods can recover high value metal elements (e.g. Li, Co, Ni), but still require synthesis of new cathode from a mix of virgin and recovered materials. Lithium iron phosphate (LiFePO₄) has been selected for study as a representative cathode material due to recent mass adoption and limited economic recycling drivers due to the low inherent cost of iron. Refunctionalization of EOL LiFePO₄ cathode was demonstrated through electrochemical and chemical lithiation methods where the re-lithiated LiFePO₄ regained the original capacity of 150–155 mAh g⁻¹. The environmental impact of the new recycling technique was determined by comparing the embodied energy of cathode material originating from virgin, recycled, and refunctionalized materials. The results demonstrate that the LiFePO₄ refunctionalization process, through chemical lithiation, decreases the embodied energy by 50% compared to cathode production from virgin materials.

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1. Introduction

Energy storage devices are viewed as an integral part in enabling sustainable energy technology adoption, particularly, in transportation and renewable energy technologies such as solar and wind [1–3]. Affordable and high performing energy storage technologies can enable increased electrification of vehicles, improving overall efficiency and reducing demand for fossil fuel energy sources [4–6]. The intermittent nature of most renewable energy

technologies naturally requires the use of energy storage to load level and maintain consistent power [7–9].

Many energy storage technologies exist, however, the most prevalent technology predicted to have a significant impact on portable applications, such as electric vehicles, are lithium ion batteries [9]. The advantage of lithium ion batteries is the high energy and power densities compared to lead-acid or nickel metal hydride technologies. The use of lithium ion batteries in portable consumer electronics has become nearly ubiquitous and significant adoption is predicted in electric vehicle and renewable energy storage [10]. Worldwide sales of lithium ion batteries are estimated to nearly triple in the next decade and surpass lead acid sales [11].

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This will lead to an increase in lithium ion batteries that are produced and eventually enter the waste stream. Therefore, end-of-life (EOL) management is a critical concern warranting studies today [12].

To understand the impacts toward sustainability, the full life-cycle impacts must be taken into account. The life-cycle of lithium ion batteries follows that of most products: synthesis and use of materials to produce the cells and packs, use phase in a device, and ultimate end-of-life (EOL) [13]. Most life-cycle studies on batteries used in electric vehicles have focused on quantifying use phase impacts and improving device performance and efficiency, a response to finding that as much as 75% of the life-cycle energy has been shown to be expended during the use phase [14–17]. However, concerns about material scarcity, processing impacts, and environmental opportunities of recycling highlight the need for more extensive study on the downstream end-of-life tradeoffs in a battery life cycle [12,18–21].

Traditional recycling techniques involve smelting or leaching processes to recover valuable metals and materials that could potentially be used to synthesize new active materials [22–24]. Cathode materials are of particular interest, as they make a high percentage of the total battery mass and cost, and contain potentially critical metals (e.g. Li, Ni, Co) [25]. To reduce cell cost, lithium ion cathode chemistries with inexpensive elemental materials (e.g. Fe, Mn) are becoming more prevalent. However, the potential secondary value of these cathode materials recovered by traditional recycling techniques is also correspondingly reduced, thus decreasing the economic incentive for recovering these materials [26]. For example, the value of the recoverable LiFePO₄ materials through traditional recycling techniques is on the order of \$0.75/lb whereas the cost of the cathode itself is more than an order of magnitude higher at \$9.10/lb [27]. The EOL management of these cells must be proactively addressed to provide an economic incentive alongside the environmentally motivated policies such as battery disposal bans (e.g. 2012 New York state ban).

As opposed to traditional recycling methods, cathode refunctionalization offers the potential for greater environmental and economic savings and may potentially improve the use phase performance [28]. As previously defined, refunctionalization is the treatment of active materials in order to regain electrochemical performance at EOL [28]. The ability to systematically implement such a technology, however, depends on demonstration of both technical feasibility as well as environmental and economic benefits. This work investigates the ability to regain the capacity of a lithium iron phosphate (LiFePO₄) cathode using a refunctionalization treatment based on re-lithiation techniques. Re-lithiation has been suggested in recent work as a possible EOL treatment for a different cathode chemistry (LiMn₂O₄) using lithium carbonate [29]. However, the present study is the first to demonstrate electrochemical performance data in the recovered LiFePO₄ cathode materials after electrochemical and chemical lithiation of EOL LiFePO₄ from commercial cells. In addition, the embodied energy of the refunctionalization process (lithiation) was compared to LiFePO₄ produced from virgin and recycled materials.

2. Experimental

2.1. 18650 Cycling

Commercial lithium ion batteries in an 18650 cylindrical form factor (18 mm diameter × 65 mm length) containing high power LiFePO₄ cathode chemistries were obtained by dismantling power tool battery packs to isolate individual cells. Cells were extracted from an 18 V Dewalt XRP NANO™ phosphate pack by manually separating cells from the pack. These cells are manufactured by

Sony Corporation with markings SE US18650FT and have a rated capacity of 1.1 Ah and a nominal voltage of 3.2 V. Each power tool pack was purchased new from the Home Depot to obtain as-purchased cells that were most likely subjected to initial formation cycles. Individual 18650 cells were cycled galvanostatically using an Arbin BT-2000 from 3.6 to 2.0 V at a 2C charge rate (2.2 A) and discharged at a 5 C rate (5.5 A) in a 60 °C convection oven to accelerate capacity fade. After 3000 cycles, the cells reached a capacity fade resulting in 80% of the original measured capacity which was used as the end-of-life (EOL) state in this study.

2.2. 18650 Disassembly and electrode electrochemical testing

All cells were removed from pack, and the Sony 18650 cells were fully discharged to 0.5 V and disassembled to obtain electrodes both directly after removal from the power tool pack (as-purchased cells) and after cycling to 80% of the original capacity (EOL cells). The 18650 cylindrical cells were dismantled in an Argon filled glove box using a pipe cutter to remove the cells vented cap and a micro flush cutter to remove the rest of the can. The 18650 “jelly-roll” was then unrolled and the cathode, anode, and separator were isolated. The cathode and anode from both the as-purchased and EOL cells were then tested in a 2032 coin cell vs. Li foil to determine the specific capacity of the electrodes. The coin cells were cycled at a C/10 rate (0.11 A) from 3.6 V to 2 V with a 1.2 M LiPF₆ EC:EMC electrolyte using a constant voltage step at 3.6 V on the charge until the current dropped below 10% of the C/10 value. Thermogravimetric analysis (TGA) was used to measure the cathode composite decomposition and estimate the specific mass of materials within the composites. Fresh anodes were coated to pair with as-purchased and EOL cathode coatings to determine electrochemical performance in a full coin cell without the availability of excess lithium. The fresh anode was capacity matched to the cathode coatings. The slurry contained a mixture of 91.5(active graphitic carbons): 8 (Polyvinylidene fluoride): 0.5 (carbon black—Super P) w/w% in N-Methyl-2-pyrrolidone (NMP) and was coated using an adjustable blade coater onto 20 μm thick copper foil. The coating was dried on the coater at 80 °C for 1 h and was further dried in a vacuum oven at 100 °C for 1 h. The anode was then paired with the recovered as-purchased and EOL anode in a 2032 coin cell and electrochemically tested in the same manner as the previous coin cells.

2.3. Electrochemical and chemical lithiation of EOL LiFePO₄

The ability to recover the electrochemical performance of the EOL cathode was investigated through electrochemical and chemical lithiation techniques. EOL cathode coatings were *electrochemically lithiated* by cycling the cathode coating vs. pure lithium in a coin cell using the method described in Section 2.2, stated previously, for several cycles and then discharging the coin cell to fully lithiate the cathode. The coin cell was then disassembled using a coin cell disassembling tool (Hohsen) in an Argon filled glove box to obtain the “re-lithiated” cathode. The electrochemically lithiated cathode was then paired vs. a fresh anode to compare to the as-purchased cathode in a coin cell and electrochemically tested using the method described in Section 2.2.

Chemical lithiation of the EOL cathode was done by scraping the cathode coating from the aluminum current collector and grinding the powder using a mortar and pestle. The ground EOL cathode coating was placed in a 1 M solution of lithium iodide (LiI) in acetonitrile and stirred for 20 h to chemically lithiate the LiFePO₄ cathode. The cathode materials were recovered through vacuum filtration using a 0.1 μm polypropylene filter paper and rinsed with acetonitrile to remove excess LiI. The recovered cathode materials

were dried under vacuum at 100 °C for 1 h and ground with a mortar and pestle. A composite with the recovered materials was prepared by mixing in NMP and coating onto aluminum foil following the same method described in Section 2.2 in casting the fresh anode. The chemically lithiated composite was tested electrochemically in a 2032 coin cell versus both Li/Li⁺ and a capacity matched fresh anode.

2.4. Materials characterization

The as-purchased, EOL, and chemically lithiated cathode materials were characterized using scanning electron microscopy (SEM) and x-ray diffraction (XRD). SEM was performed using a field emission Hitachi S-900 microscope at 2 kV. XRD measurements were performed with a D2 Phaser benchtop XRD system (Bruker AXS, Germany) using Co K α radiation ($k = 1.789 \text{ \AA}^{-1}$) scanning between 20° of 10° and 80°. All samples were prepared by uniform packing of material in sample holders.

2.5. Embodied energy analysis

The metric of cumulative energy demand (CED) was applied to determine the environmental tradeoffs of producing new virgin materials as compared to a refunctionalized cathode. CED includes all energy inputs across the life cycle of the materials, including both direct process electricity, as well as energy required to produce electricity and other raw materials. As a baseline, the CED was calculated for synthesis of LiFePO₄ from virgin materials using existing LiFePO₄ hydrothermal production inventory data [16] and calculated using the CED method in SimaPro (Pé, The Netherlands). The LiFePO₄ chemicals and energy inventory was modeled using ecoinvent v3 as unit processes.

The CED of producing LiFePO₄ from recycled materials was modeled using intermediate physical recycling and re-synthesis of LiFePO₄ using a recycled lithium source. Intermediate physical recycling involves shredding and separation of the lithium containing components which are converted to lithium carbonate to synthesize new cathode materials. The CED to recover the lithium carbonate has been modeled in previous work [30] to be 2.6 mmBtu/ton of recovered lithium carbonate. The recycled lithium carbonate is used to produce the lithium hydroxide precursor in the hydrothermal synthesis. Therefore, the CED for the recycled lithium carbonate is equal to the intermediate recycling CED, and eliminates upstream impacts from the production of virgin lithium carbonate. The recycling of lithium carbonate was modeled using ecoinvent v3 inventory unit processes data as well.

The CED to refunctionalize EOL LiFePO₄ cathode through chemical lithiation was modeled using direct physical recycling to recover the EOL cathode, and the energy and materials consumed in the chemical lithiation process. Direct physical recycling involves only shredding and filtering processes to isolate the EOL cathode material and has been modeled in previous work [30] to be 2.34 mmBtu/ton of cathode material recovered which was used in this calculation. Chemical lithiation and refunctionalization energy impacts were modeled based on the re-lithiation of 1 kg of EOL LiFePO₄. The amount of lithium needed for re-lithiation of 1 kg of EOL LiFePO₄ was calculated using the capacity loss of the EOL cathode and multiplying by lithium capacity (3860 mAh g⁻¹). Stoichiometric ratios were used to calculate the mass of LiI required, and the resulting mass of acetonitrile to make a 1 M solution. LiI production CED was modeled from the reaction of lithium hydride with iodine in diethyl ether [31] using GaBi software and ecoinvent v3 database. A solvent recovery CED of acetonitrile and the diethyl ether used in the LiI production were calculated following previously published procedures [32] and GaBi

software and ecoinvent v3 database as well. Direct measurements (Watts up? PRO ES AC power meter) and nameplate equipment power consumption were used to model the energy required to stir, filter, and dry the material at the lab scale. In all cases, the energy consumption of the synthesis or refunctionalization process was modeled using U.S. medium voltage electricity mix.

3. Results and discussion

3.1. As-purchased vs. EOL electrodes

Commercial 18650 cells containing a lithium iron phosphate (LiFePO₄) cathode chemistry were obtained from a power tool pack shown in Fig. 1 inset. Some of the individual cells were then subjected to accelerated cycling at 60 °C at a 2C charge and 5C discharge rate to force a capacity fade in a reasonable amount of time. After 3000 cycles (approximately 3 months), the cycled cells had faded to 80% of the original measured capacity and were used as end-of-life (EOL) cells in this study. The cycling performance of one of the cells is demonstrated in Fig. 1. This capacity fade is similar to recent work on LiFePO₄ cell cycling [33].

The EOL and as-purchased (not cycled) cells were disassembled to obtain the individual electrodes for testing versus lithium to determine the performance of the isolated electrodes. The first cycle voltage curves for the as-purchased and EOL cathodes are shown in Fig. 2a. The first cycle charge capacity for the EOL LiFePO₄ active material is significantly lower, 110 mAh g⁻¹, than the charge capacity for the as-purchased LiFePO₄ active material, 157 mAh g⁻¹. However, the discharge capacity for the two cells is in the expected capacity range for LiFePO₄ of 150–155 mAh g⁻¹. During charge, lithium ions are being removed from the cathode structure; therefore, a lower charge capacity indicates a loss of lithium from the structure and is a main contributor to the full cell capacity fade. The increase in capacity upon discharge demonstrates that the LiFePO₄ structure may be able to re-lithiate electrochemically versus a lithium metal anode. The as-purchased cathode has a charge capacity much closer to that of the discharge capacity demonstrating that the as-purchased cathode has a fully lithiated LiFePO₄ structure.

The difference in crystal structure of the cathode was further investigated using x-ray diffraction (XRD). The as-purchased and EOL cathode composites were scraped from the aluminum current collector and ground in order to take powder XRD measurements.

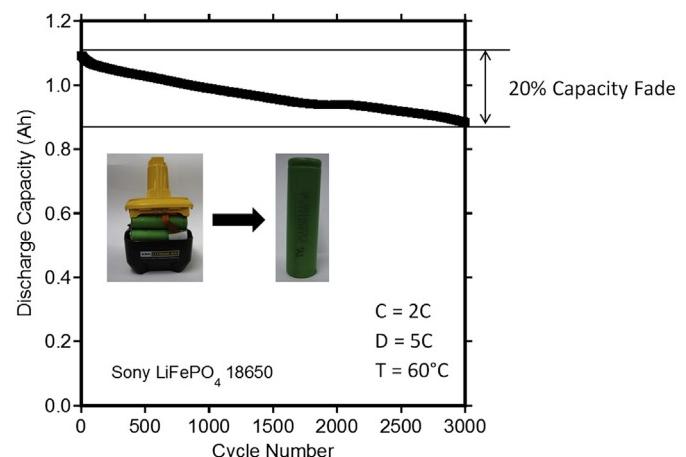


Fig. 1. The electrochemical cycling performance of an 18650 cell under accelerating testing conditions. (Inset) Pictures of partially disassembled power tool pack containing 18650s with LiFePO₄ cathode chemistry.

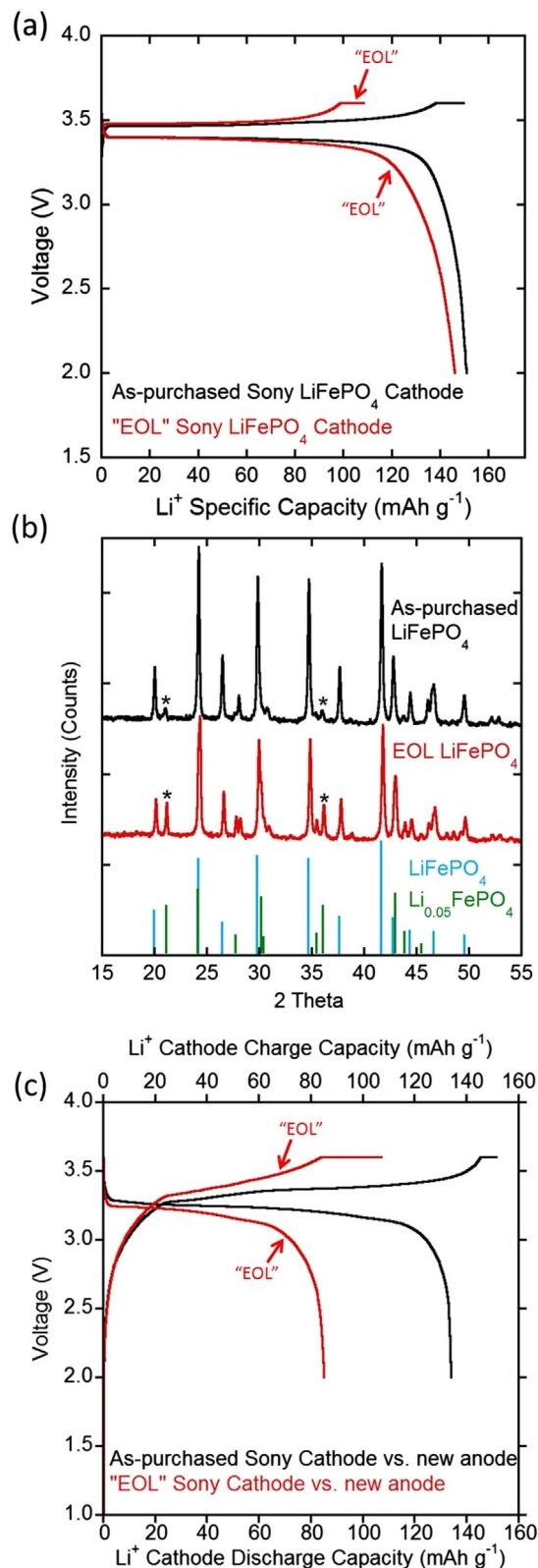


Fig. 2. (a) Voltage profiles for as-purchased and EOL LiFePO₄ cathodes vs. Li/Li⁺ at C/10 rate. (b) X-ray diffraction of as-purchased (black) and EOL (red) LiFePO₄ cathodes with reference peaks for LiFePO₄ (blue-ICDD # 01-076-8586) and Li_{0.05}FePO₄ (green-ICDD # 01-075-7582). (c) Voltage profiles for an as-purchased and EOL LiFePO₄ cathode vs. a newly coated anode at C/10 rate. The electrolyte was 1.2 M LiPF₆ EC:EMC in a 3:7 ratio for all electrochemical testing. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

The XRD spectra can be seen in Fig. 2b where both cathodes demonstrate the reference LiFePO₄ peaks shown in light blue. However, an increase in intensity and appearance of new peaks is observed for the EOL cathode. These peaks marked by a star in Fig. 2b are indicative of a delithiated Li_{0.05}FePO₄ phase (reference lines shown in green) within the EOL cathode. The XRD results further support the conclusion that some of the LiFePO₄ within the EOL cathode has lost lithium from the structure due to cycling.

To test the performance of the as-purchased and EOL cathodes without the presence of excess lithium, a new anode was cast that was capacity matched to the cathodes and the electrodes were paired versus each other in a full cell. The voltage profiles for both the as-purchased and EOL cathodes versus the new anode can be found in Fig. 2c with capacity stated with respect to the cathode. The results again indicate that the EOL cathode lost lithium from some of the LiFePO₄ resulting in an initial capacity of 85 mAh g⁻¹ (consistent with charge capacity in Fig. 2a) compared to 132 mAh g⁻¹ for the as-purchased cathode. It should be noted that the initial discharge capacity is a somewhat lower than expected for the as-purchased cathode due to the first cycle loss from formation of a solid electrolyte interphase (SEI) layer on the new anode which is consistent for all samples paired with a new anode.

3.2. Electrochemical and chemical refunctionalization of EOL cathode

EOL cathode refunctionalization was investigated through electrochemical and chemical lithiation. As characterized in the previous section, the main capacity fade mechanism of the EOL cathode, in this case is due to the loss of lithium due to cycling which caused lithium to be irreversibly lost. However, after removal from the 18650, the electrochemical results for the EOL cathode vs. lithium demonstrated the ability to re-lithiate the cathode as the discharge capacity of the cell was observed to be the same as the as-purchased cathode. Therefore, the ability to refunctionalize the EOL cathode through electrochemical lithiation was investigated by cycling the EOL cathode vs. lithium in a coin cell for multiple cycles and discharging the cell to fully lithiate the cathode. The cell was then disassembled and the electrochemically lithiated cathode coating was then paired vs. a capacity matched new anode. The first cycle voltage profiles comparing the as-purchased LiFePO₄ cathode and the electrochemically lithiated EOL cathode versus the new anode are shown in Fig. 3. The results demonstrate that the cathode capacity can be regained through electrochemical lithiation where the refunctionalized cathode had a similar charge and discharge capacity compared to the as-purchased cathode. The capacity of the electrochemically lithiated cathode versus a graphitic anode was slightly higher than the as-purchased cathode which is most likely due to a small difference when capacity matching with the anode. The capacity of the as-purchased and electrochemically lithiated cathode should be viewed as equivalent and within error. The cycling performance was also found to be similar after 50 cycles as shown in Fig. 3 (right) demonstrating that re-lithiated LiFePO₄ cathode can retain the capacity of the as-purchased cathode over this range of cycles.

Although electrochemical lithiation is demonstrated to refunctionalize the EOL LiFePO₄ cathode and to enable the reuse of the cathode in a new cell, the process may not be scalable and relies on the cathode having sufficient mechanical adhesion to the current collector at EOL. An alternative technique (and potentially more scalable) is to remove the cathode from the current collector and use a solution based chemical lithiation. Therefore, chemical lithiation of the EOL cathode was investigated by scraping the cathode composite from the aluminum current collector, grinding to a

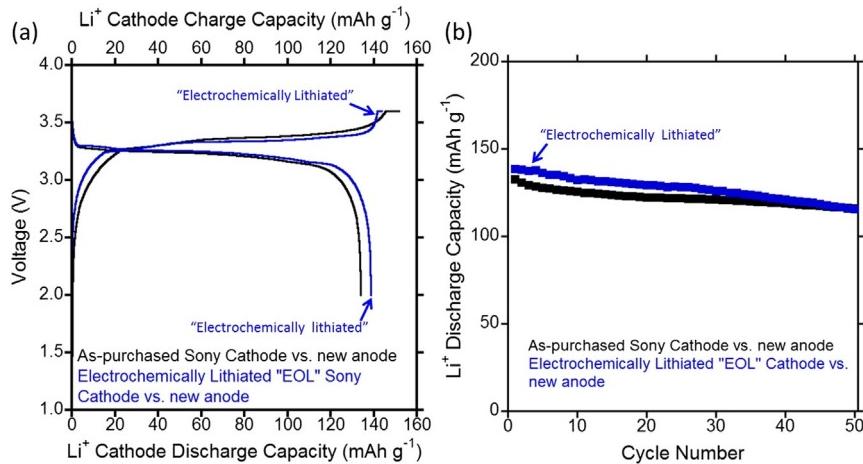


Fig. 3. (a) Voltage profiles and (b) cycling discharge capacity for an as-purchased cathode (black) and an electrochemically lithiated EOL cathode (blue) vs. a new anode at a C/10 rate with 1.2 M LiPF₆ EC:EMC in a 3:7 ratio electrolyte. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

powder and stirring in a 1 M Lithium Iodide (LiI) solution in acetonitrile for 20 h.

Immediately after addition of the cathode composite to the LiI solution, the color visibly changes from a light yellow solution to a brown solution due to the formation of Iodine in the solution. XRD analysis was performed on the chemically lithiated cathode composite as shown in Fig. 4. The data for chemically lithiated LiFePO₄ does not exhibit de-lithiated peaks associated with delithiated LiFePO₄ (where the location of peaks are starred), but instead has a similar spectrum to that of the as-purchased LiFePO₄.

Scanning electron microscopy images were acquired for as-purchased, EOL, and chemically lithiated cathode composites scraped from the aluminum current collector and are shown in Fig. 5a, b, and c respectively. The as-purchased and chemically lithiated images do not show any qualitative differences between the two samples whereas the EOL sample has the presence of a greater amount of smaller particles, possibly due to the formation of excess FePO₄ with the loss of lithium. The images show the presence of the cathode particles as well as high aspect ratio conductive carbons (i.e. possibly VGCF or MWCNTs) which can be

found in all images. This indicates that the conductive additive was not inadvertently removed during the chemical lithiation and filtration process.

The chemically lithiated cathode composite was dispersed in N-Methyl-2-pyrrolidone (NMP) and coated onto an aluminum current collector. The coating was well-adhered to the current collector which indicates that the polymer binder in the cathode composite was retained through the chemical lithiation process as well. The chemically lithiated cathode composite was tested versus lithium and a capacity matched new anode and the voltage curves can be seen in Fig. 6a and b respectively. The results demonstrate that the chemically lithiated cathode had a discharge capacity very close to that of the as-purchased sample for both cases. A larger first cycle loss was determined when tested versus a graphitic anode due to a higher first cycle charge capacity for the chemically lithiated sample which may be due to excess lithium retained after the chemical lithiation process. However, the similar discharge capacity and material characterization demonstrate that both chemical and electrochemical lithiation are promising new methods to reuse active materials through material refunctionalization while retaining the conductive additives and binder.

3.3. Cumulative energy demand (CED) analysis

The economic and environmental benefit of refunctionalization compared to synthesis from virgin or traditionally recycled materials is owed to the ability to recapture valuable active materials before they are returned to a lower value elemental level. Of course, the cost and energy intensity of the recovery and isolation of the cathode material (or composite) and refunctionalization process must be taken into account as well. To quantify potential tradeoffs in recovery or recycling methods, cumulative energy demand (CED) was calculated for each potential EOL pathway and compared to the initial impacts of producing the virgin raw materials. Intermediate recycling is modeled for the production of LiFePO₄ from a recycled lithium source. A lithium carbonate source is recovered through shredding and precipitation and the energy of the process has been modeled in previous work [30]. The present results (Fig. 7) demonstrate a reduction of approximately 10% in embodied energy when cathode materials are recycled and then used in the synthesis of a new cathode, as compared to the all virgin raw material baseline. In the case of the refunctionalization, physical separation has a higher CED than traditional recycling due to the energy to recover all 1 kg of EOL LiFePO₄ as compared to the allocation of

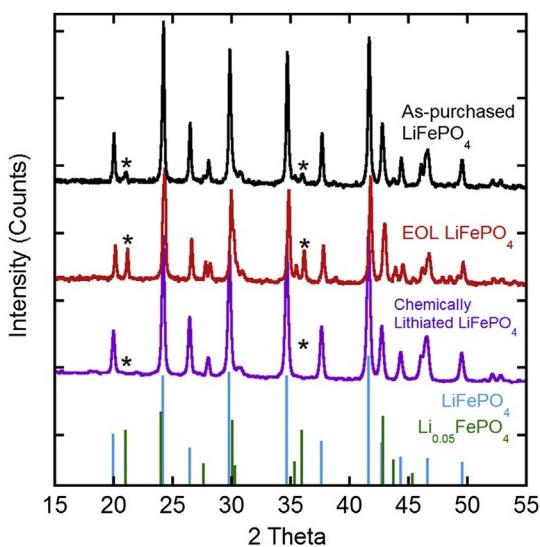


Fig. 4. XRD of as-purchased, EOL, and chemically lithiated LiFePO₄ cathode composites.

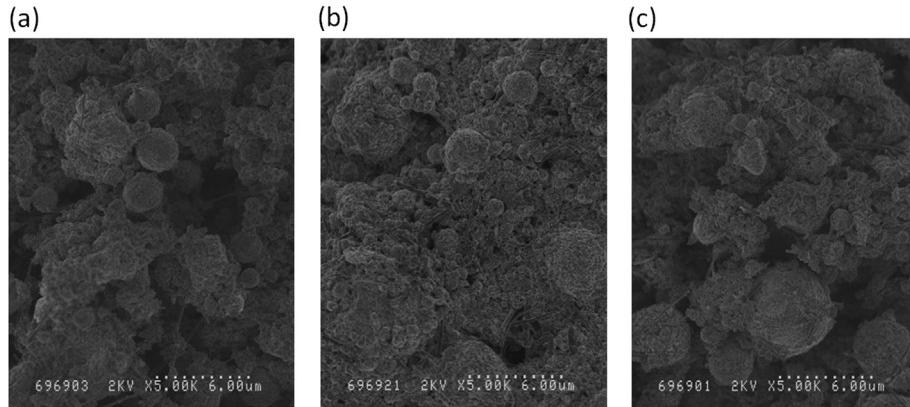


Fig. 5. Scanning electron microscopy (SEM) images of (a) as-purchased (b), end-of-life and (c) chemically lithiated LiFePO₄ cathode coatings scraped from Al current collector.

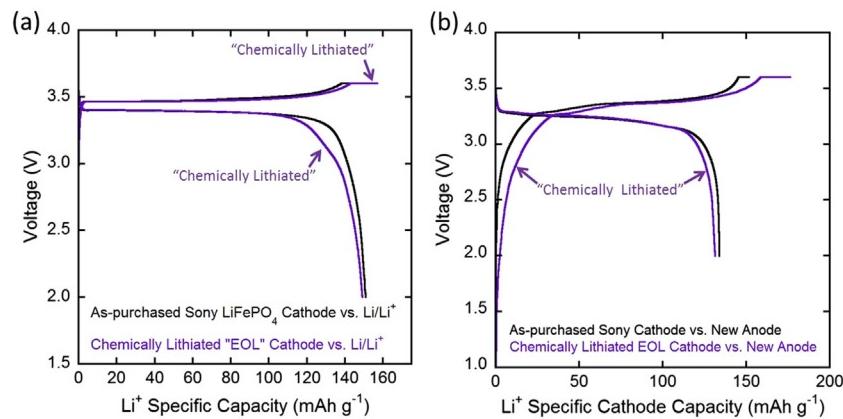


Fig. 6. Voltage profiles for as-purchased (black) and chemically lithiated EOL (purple) LiFePO₄ cathodes versus lithium (a) and a capacity matched new anode (b). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

energy to recover just the lithium source. However, the net CED is reduced by 50% due to the complete elimination of any virgin material demand and a re-lithiation process that uses low temperature and minimal energy. It should be noted as well that the chemical lithiation processes were modeled at the lab scale and

further CED reduction is anticipated with scale up. Overall, refunctionalization represents a viable pathway to handle end of life battery materials with a lower CED and presents another option for EOL management with lithium ion batteries.

4. Conclusions

Commercial LiFePO₄ containing cathodes were cycled to an EOL state and recovered from the cylindrical 18650 cells. Coin cell testing and x-ray diffraction revealed that the main fade mechanism is caused by a loss of lithium during cycling. The ability to refunctionalize the EOL cathode to as-purchased performance was demonstrated using electrochemical and chemical lithiation. These results offer a method to reuse lithium ion cathode materials at EOL which can reduce cost and energy of synthesizing new cathode materials. The potential environmental benefit of the chemical lithiation process was confirmed by calculations of cumulative energy demand, showing a 50% reduction in CED compared to synthesis of virgin materials.

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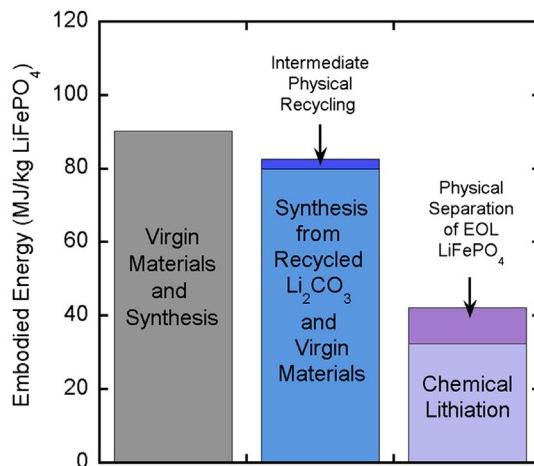


Fig. 7. Embodied energy for LiFePO₄ produced through synthesis using virgin materials (gray) and recycled materials (blue), and chemical lithiation of EOL LiFePO₄ (purple). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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